UNIVERSITY OF PENNSYLVANIA

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GEORGE HERBERT BANCROFT

A DISSERTATION

IN PHYSICS

PRESENTED TO THE FACULTY OF THE GRADUATE SCHOOL IN

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Intensity Variation of L Series X-Ray Lines with Tube Voltage

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The variation of intensity of the L series x-radiation, from a copper target, with the voltage applied to the x-ray tube has been studied. The voltage range over which this investigation was made included the K excitation value for copper. A definite increase was found in the slope of the intensity-applied voltage curve, which appeared immediately after the K state was excited. The intensity was plotted against the square of the voltage difference between the K excitation and the applied voltages, and the resulting curve was found to be substantially a straight line. This result is in accord with the theoretical predictions of Smekal and is an improvement over the experimental work of Stumpen.

SMEKAL[1] has pointed out, on a theoretical basis, that the intensity of x-radiation of any series should show increases as each harder series is excited successively by increasing the voltage on the tube. Experimental investigations of this prediction have been carried on by G. Kettmann,[2] A. Jönsson,[3] and H. Stumpen,[4] of whom the latter has done the most extensive work. Kettmann,[2] working on lanthanum L_α and L_β radiation, employed the photographic method, comparing the intensities obtained at different voltages by the use of a photometer. He was unable to find positive evidence of the increase in intensity predicted by Smekal.[1] A. Jönsson[3] used a Geiger point counter to measure the intensity variation of the L_α and L_β lines of molybdenum. The maximum value of the exciting voltage was 21 kilovolts. Since the K critical voltage for molybdenum is 19.9 kilovolts, Jönsson[3] obtained only two points on the line intensity tube voltage curve above the critical voltage value and the slope of the line passing through the two points did not depart obviously from that for points lying below. H. Stumpen,[4] using the ionization method for measuring intensities, studied the intensity variation with voltage of the tungsten L_β and L_γ lines. His curves showed a gradually increasing slope beginning at about 70 kilovolts whereas the K excitation voltage for tungsten is 69.9 kilovolts. Further, he reported that the intensity above this point increased linearly with the square of the applied voltage. These facts are summarized in Table I. Compton and Allison[5] say, in their discussion of this work, that, "Although such an effect was predicted by Smekal,[1] Kettmann[2] failed to find positive evidence for it and experimentally its existence does not seem to be satisfactorily demonstrated."

The work of E. Lorenz,[6] on the voltage intensity relation for the aluminum K series showed that no such increases in intensity were to be expected where there was no possibility of exceeding the critical potential of a harder characteristic radiation. Theories of collision ionization, such as those of Davis,[7] Rosseland,[8] Thomas,[9] Webster[10] and others[11] have been limited to the investigation of the direct excitation of a single level by electron impact; hence they could not be expected to offer any explanation of an increase in intensity connected with the critical potentials of harder characteristic radiation.

The investigation described in this paper was undertaken to obtain additional experimental evidence concerning the behavior of the intensity of a group of x-ray lines of the L series when the critical K voltage of the same element was exceeded. The method used to estimate these

[1] A. Smekal, Verh. d. D. Phys. Ges. **21**, 149 (1919).
[2] G. Kettmann, Zeits. f. Physik **18**, 359 (1923).
[3] A. Jönsson, Zeits. f. Physik **43**, 845 (1927).
[4] H. Stumpen, Zeits. f. Physik **36**, 1 (1926).
[5] Compton and Allison, *X-Rays in Theory and Experiment* (D. Van Nostrand Co. 1935), p. 81.
[6] E. Lorenz, Zeits. f. Physik **51**, 71 (1928).
[7] Bergen Davis, Phys. Rev. **11**, 433 (1918).
[8] Rosseland, Phil. Mag. **45**, 65 (1923).
[9] Thomas, Proc. Cam. Phil. Soc. **23**, 829 (1927).
[10] Webster, Hansen, and Duveneck, Phys. Rev. **43**, 839 (1933).
[11] Reference 5, page 73.

TABLE I. *Variation of intensity with tube voltage.*

RADIATION STUDIED	MAXIMUM VOLTAGE	METHOD OF INTENSITY MEASUREMENT	VOLTAGE SUPPLY	AUTHOR
Lanthanum L_β L_α	1.03 V_k=39.9 kv 1.08 V_k=41.9 kv	Photographic	Induction machine	Kettmann[2]
Tungsten L_β L_γ	1.323 V_k=90.0 kv 1.323 V_k=90.0 kv	Ionization Ionization	500-cycle a.c. Unrectified	Stumpen[4] Stumpen[4]
Molybdenum L_α L_β	1.055 V_k=21.0 kv 1.055 V_k=21.0 kv	Geiger Point Counter	500-cycle a.c. Rectified and filtered	Jönsson[3]

intensities, was to measure the photoelectric current obtained from an insulated metal plate, which was placed in the path of x-rays. The number of photoelectrons ejected from the surface per second was proportional to the number of x-ray quanta striking it in that time interval.

THE EXPERIMENTAL ARRANGEMENT

The high voltage for the x-ray tube was obtained by transforming the regular 60-cycle a.c. supply. The high voltage thus obtained was rectified by the use of a full wave, four kenotron, bridge circuit the output of which was further smoothed by two 0.1 microfarad condensers. A resistance of 50,000 ohms was used in series with the x-ray tube to reduce the effect of any unavoidable arc due to the liberation of gas. The voltage was measured in terms of the current flowing through ten, one megohm, Shallcross, precision, wire wound resistors connected in series.

The x-ray tube was mounted directly on the spectrograph as shown in Fig. 1, and was sealed in position with sealing wax. No window was used between the x-ray tube and the spectrograph, both of which were separately connected to the pumping system, which was of a standard type. Electrode holders, provided with sylphon adjustments *H–H*, were fitted to the ground ends of the Pyrex separator tube as shown in the figure. These adjustments provided a limited motion for each of the electrodes back and forth along the axis of the tube and thus made it possible to bring the focal spot and the collimating system of the spectrograph into accurate alignment. The motion of the cathode made possible a limited external control of the size and uniformity of the focal spot. The actual cathode was of the indirectly heated, oxide-coated type. Around this cathode a focusing shield was placed. By adjusting the distance between the surface of the cathode and this shield almost any degree of focusing could be obtained. The anticathode was water cooled and was provided with an internal baffle which constrained the water to flow smoothly and continuously across the rear surface of the target face.

A small, plane, ruled glass grating, having 2406 lines per inch was used. The ruled surface was approximately 2.5×8 mm, and the grating was mounted so that the axis of rotation passed along the ruled surface. A knife edge, which was brought up quite close and parallel to the ruled surface, formed the second slit of the collimating system of the spectrograph. This slit was 0.220 mm in width, whereas the first slit was 0.275 mm wide. The last slit S_3, which selected the radiation desired, was situated at a point about 15 cm from the grating and was 0.990 mm wide. Immediately behind this last slit, a small nickel vane *B* was introduced through the wall of the spectrograph by the use of a ground glass joint, in such a way, that it could be set parallel or perpendicular to the x-ray beam, thus either allowing the beam to pass or cutting it off completely from the rest of the spectrograph chamber. This vane was electrically connected to the grounded case of the spectrograph. The purpose of the vane was to allow the comparison of the position of the galvanometer spot, with and without the x-ray beam affecting the measuring circuit. An ion trap *C* was arranged as shown

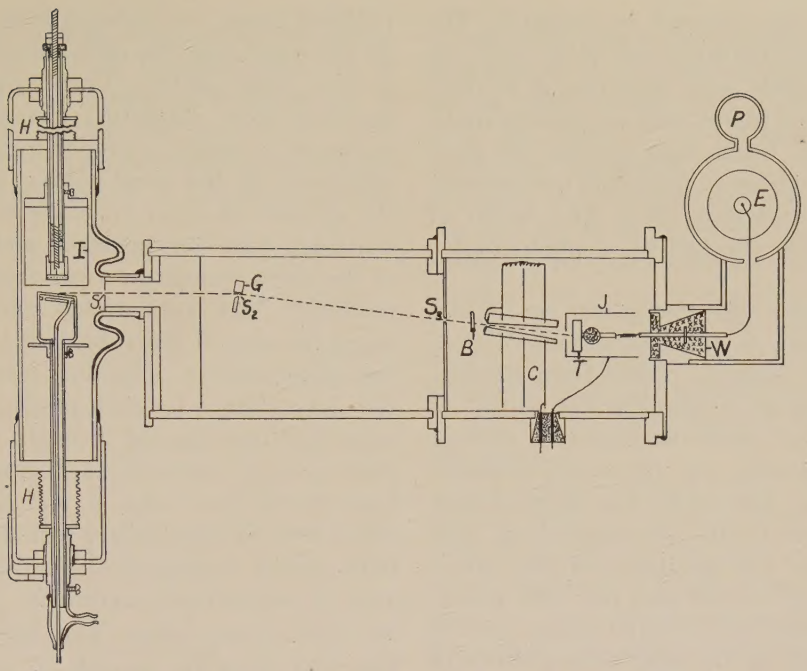

FIG. 1. A diagram of the x-ray tube, vacuum spectrograph, and detecting device used.

in Fig. 1, between the vane B and the rest of the chamber. The almost parallel plates through which the x-ray beam passes were purposely made to diverge in the direction away from the source in order both to prevent deflection of the beam by reflection and to minimize the generation of photoelectrons. A potential difference of about 90 volts was maintained between these plates, as is indicated in the figure. Having passed through the ion trap, the x-rays struck an insulated metal plate T, having an aluminum surface, and photoelectrons were produced. These photoelectrons were then swept out of the area by the field from a positive electrode J, and the insulated plate charged up positively as more and more electrons were lost. The charge from the insulated plate was conducted out of the spectrograph through the sealing wax plug W in the back plate. From this point the charge was carried to the control grid E, of the Western Electric electrometer tube D96475, by means of a semi-rigid conductor passing through brass shields with air as the insulator.

The electrometer tube was used in a Barth[12] circuit as modified by Pennick.[13] The galvanometer read the difference in voltage between the plate and the screen grid of the tube, produced in response to changes in the potential of the control grid. The circuit was tested for linearity of voltage response by the application of a number of known voltages, obtained from a Leeds and Northrup student potentiometer, calibrated in the usual manner, and it was found to be accurately linear for voltage changes as great as 0.04 volt. A rough potentiometer system P was permanently connected in the circuit between the ground and the high grid resistance, which made it possible to check the sensitivity of the circuit from time to time.

EXPERIMENTAL PROCEDURE

In the adjustment of the spectrograph, the position of the anticathode was first set visually, with the help of a carbon arc light focused upon the target at the place where the focal spot would later be formed. Then a series of photographic exposures were made with the x-rays, in which the position of the anticathode and the angle of the grating were separately adjusted until the most desirable diffraction pattern was obtained. The copper L group was identified

[12] Barth, Zeits. f. Physik **87**, 399 (1934).
[13] Pennick, Rev. Sci. Inst. **6**, 115 (1935).

and the slit S_3 was adjusted to isolate it. The ion trap and the photoelectric plate with its accompanying anode were then placed in position. Photographic plates were exposed to make certain that the beam was passing by each new obstruction without change and was actually striking the photoelectric plate. The action of the vane was also checked by a photographic method.

The electrometer tube circuit was set up and adjusted according to the instructions of Pennick.[13] The plate voltage, picked from the filament circuit, was set so that the flat portion of the galvanometer deflection-filament current curve fell accurately, at the rated filament current (270 ma). The circuit was supplied with 24 volts from four 6 volt, 120 ampere-hour, lead storage batteries. The sensitivity of the circuit was approximately 15,000 mm per volt, which, with a grid resistor of 9.6×10^{11} ohms, gave a current sensitivity of the order of magnitude of 6.9×10^{-17} amperes per mm. The circuit was found to have a period of three minutes for full deflection, but on investigation of the time rate of deflection, it was found that, for the magnitude of the deflection usually obtained, 85 percent of the total movement took place in the first minute. Also, if the time interval were carefully controlled, the deflections at the end of a minute were reproducible, and it was therefore considered legitimate to read the deflections after the x-rays, in each case, had been falling on the plate for one minute. The time necessary to take a full curve was thus reduced to three hours. This shorter time was advantageous since the possibility of keeping the conditions in the x-ray tube reasonably constant while a curve was being observed was thus increased. The current through the x-ray tube was kept constant throughout an observation to within 0.2 ma or 2 percent, by manipulation of a resistance in the cathode heater circuit. It was found that more stable operation was obtained when the voltage on the x-ray tube was immediately brought up to its maximum value and then reduced in the steps desired to obtain the intensity voltage curve.

After a few hours of operation the anticathode surface became roughened. The anticathode was then removed and its surface was either polished down, or replaced if it were too thin. At the same time the cathode was removed and given a fresh coating. Each time the electrode positions were disturbed a readjustment was necessary to assure a proper focus and a proper alignment of the focal source with the entire slit system. In order to make this adjustment, the x-ray tube was operated and the deflection of the galvanometer at a particular voltage was recorded. The anticathode was then moved along the axis of the tube in 0.1 mm steps and the galvanometer deflections were observed at the same voltage for each position until a maximum had been passed through. At this adjustment a small piece of photographic plate was fastened to the vane B and a photographic check was obtained. Care was always taken to have a fresh coating on the cathode and a fresh area of anticathode surface for the focal spot, for each curve taken. The cathode structure was just eccentric enough to move the focal spot to a new position if it were rotated through 180°. The separation of the focal spots thus obtained was as much as two or three mm. In this way it was possible to use the same polished surface of the anticathode to obtain two curves.

Results

The spectrograph, as used, had a rather low dispersion and the whole group of copper L lines was selected by the final slit, instead of only one line of the group. However, if the intensity of the L lines is to be increased due to the emission of K radiation, according to the Kossel hypothesis, then those lines arising from vacancies in either the L_{II} or the L_{III} level should experience an increase in intensity and the total increase should be greater than if only a single line were observed. It was, consequently, thought best to use the entire L group for the intensity measurements.

A number of curves, of which those in Fig. 2 are typical, were obtained, showing the behavior of the intensity with increasing voltage, in the neighborhood of the K critical potential. Each point on the curves represents the average of six separate evaluations of the intensity at that voltage. In Fig. 2 the individual values are shown by the small dots and the average values by the

dots with circles around them. All of the curves exhibit a definite increase in slope above the K excitation point. If the intensities above the K critical potential are plotted against $(V-V_k)^2$, the resulting curves are substantially straight lines, as may be seen in Fig. 3. If these intensities

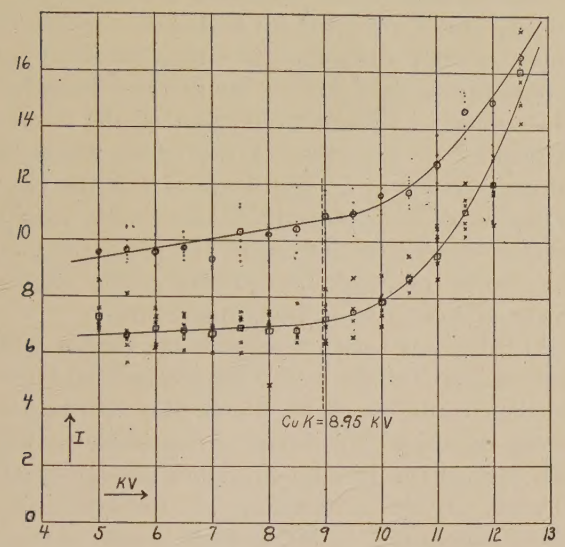

FIG. 2. The intensity of the group of Cu L lines plotted directly against the voltage applied to the x-ray tube. The ordinates represent the galvanometer deflection in cm.

are plotted against the square of the voltage applied to the tube, the resulting curves are definitely not straight lines. If it is assumed that the increase observed is due to atoms left ionized in the L state after the emission of K radiation, then one should expect that this increase would follow the same law as is obeyed by the K radiation itself. Since the law obtained from the curves in Fig. 3 agrees very well with this theoretical expectation, one may reasonably conclude that the portion of the intensity due to direct collision ionization remains roughly constant over the region investigated, which lies above the K excitation potential.

The effect of a change in the distribution of the intensity of the electronic bombardment of the focal spot was shown by a curve (see Fig. 4), taken in an effort to repeat a set of observations, after varying the voltage from the maximum to the minimum value, by changing it in the reverse direction from the minimum back to a maximum again. Between six and seven kilovolts on the return curve a sharp break occurred after which the course followed was approximately parallel to that taken in the original set of observations and was displaced from it in the direction of reduced intensity. This break was correlated with a quite noticeable change in the character of the focal spot. During the first set of observations the focal spot possessed three quite definite areas and, at the break in the curve, one of these areas was seen to disappear. During the remainder of the repeat curve the character of the focal spot remained constant. In accordance with the results of this test all curves showing irregularities were discarded.

The curves obtained (Fig. 2) show increases of intensity over that at the critical voltage varying from 54 percent to 128 percent over the voltage range employed. This increase appears to begin close to the critical voltage for K excitation. The curves also seem to have flattened out somewhat, before ten times the L critical potential of 950 volts is reached as judged from the portion of the curve below the K critical value. This seems to be in qualitative agreement with the results of Lorenz on the Al K_α line.

The results reported in this paper agree with those of Kettmann,[2] Stumpen,[4] and Jönsson,[3] insofar as they indicate an increase in the intensity of the L group when the K excitation voltage is exceeded. However, they are in definite disagreement with those of Stumpen[4]

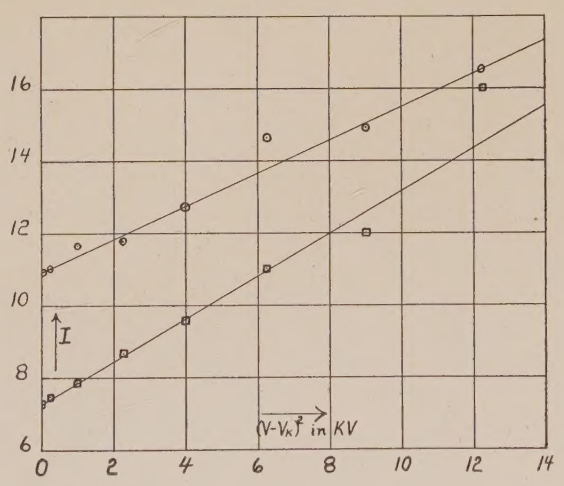

FIG. 3. The intensity of the group of Cu L lines plotted against $(V-V_k)^2$. The ordinates represent the galvanometer deflection in cm.

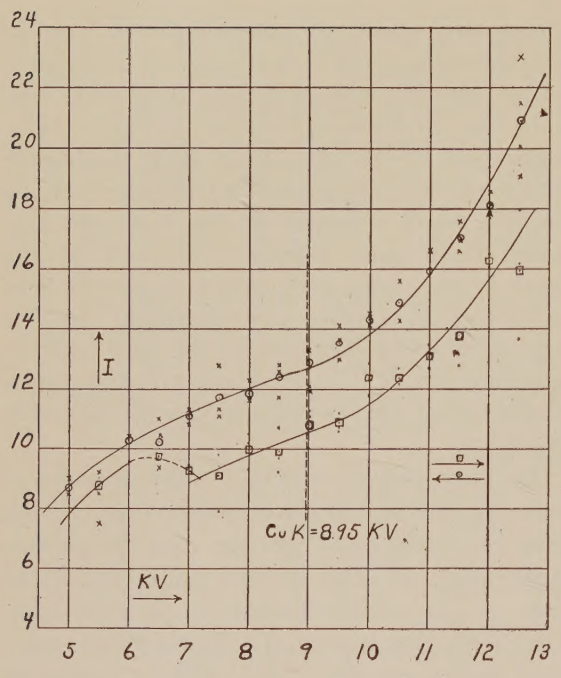

FIG. 4. The intensity of the group of Cu *L* is plotted against the voltage applied to the x-ray tube and shows the effect of a change of the distribution of the intensity of electron bombardment in the focal spot of the tube. The change coincides with the break in the curve.

in that the intensity is not proportional to the square of the applied voltage but to the square of the difference between the *K* critical potential and the applied voltage. The conditions employed in this investigation may be considered as exceptionally favorable for the determination of this relation, as the maximum in the intensity voltage curve, reported by E. Lorenz, occurs in approximately the region in which the increase in intensity begins. Hence the increase is added to a practical horizontal portion of the curve, instead of to a portion of the curve which is normally rising at a rapid rate, as was the case in the work of Stumpen.[4] These results would seem to agree very well with the predictions of the theory of Smekal,[1] concerning the return of the excited atom to its normal state.

At this point I wish to express my thanks and appreciation for the many suggestions and the continued interest of Dr. C. B. Bazzoni, under whose direction this investigation was carried out, and to the Randal Morgan Laboratory of Physics of the University of Pennsylvania, whose laboratory facilities were placed at my disposal, thus making this work possible.

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